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13. ABSTRACT (Maximum 200 words)  Over the awarded period, a new optical laboratory to characterize hole burning materials was completed. Si doped diamond films, Eu <sup>3+</sup> and Tu <sup>3+</sup> doped polymers and Tu <sup>3+</sup> doped sol-gel glasses were studied. The diatomic Si <sub>2</sub> of the defect center in diamond was established. Low temperature persistent hole burning was observed with hole width of 0.01 cm <sup>-1</sup> . Hole width dependence on irradiation dose and temperature were studied. A photoinduced hole filling was observed. A 2D imaging amplitude correlation technique was developed and demonstrated for single shot photon echo read out with a read out speed of 27 Terabits per second. Photon echo dependencies on exposure time, delay time between data and reference pulses, and number of readout cycles were studied. Up to 10 <sup>4</sup> single-shot readout cycles by a femtosecond pulse were observed without substantial echo signal degradation.		

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**Materials for Spectral Hole-Burning Storage**

May 21, 1996

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Materials are one of the key element to advance the spectral hole-burning technology for optical storage and processing. Our research is aimed mostly on new materials for low ( $T < 10$  K) and high temperature optical storage. To characterize and compare different potential hole burning materials we investigated material parameters such as crossections of optical transitions, quantum yields of hole burning, inhomogeneous and homogeneous broadening, and hole lifetimes. The temperature dependence of these parameters are important for understanding their basic science and high temperature applications. The known low temperature material, free-based octaethylporphyrin in polystyrene, with large inhomogeneous broadening was studied for time domain hole-burning storage with the purpose of demonstration storage and fast retrieval of femtosecond data signals. A new single-shot interferometric crosscorrelation technique for detection of accumulated photon echo signals in femtosecond time scale was developed and demonstrated.

Over the program period, progress was achieved in three areas: 1)laboratory, 2)samples, and 3)research.

**1. Laboratory.** A new hole burning laboratory was set up to characterize materials and determine the major spectroscopic parameters. The room for the laboratory was renovated including purchase and installation of a TMC vibration isolated optical table. Major laser equipment was purchased and installed:

- Coherent Inc. model Innova 310 Argon Ion Laser System with visible power up to 10W and PowerTrack active stabilization of the optical cavity;
- Coherent Inc. model 899-29 Autoscan II single-frequency ring laser using Ti:S or Dye including integrated wavelength meter and optics for spectral range 560-840 nm.

Detection system including lock-in amplifier, small monochromator and Ga-As photomultiplier was installed. A new Oxford Instruments variable temperature helium cryostat has been purchased and helium pump and pumping line have been installed. Those equipment allows us to perform measurements at temperatures between 1.3 and 300 K.

**2. Materials.** New materials includes Si doped chemical vapor deposited diamond films (samples are being provided by Dr. W. Phillips, Crystallume, Santa Clara, CA and Prof. F. Smith, Physics Department, CCNY), diamond-like carbon and silicon-carbon alloy films (Prof. F. Smith, CCNY), Eu<sup>3+</sup> and Tm<sup>3+</sup> doped polymers (Prof. Y. Okamoto, Polytechnic University, Brooklyn, NY ), and rare-earth doped sol-gel glasses (Prof. H. Gafney, Queens College).

**3. Research.** Our research focused on a series of measurements of absorption and fluorescence spectra, electronic state lifetimes, hole burning kinetics, spectra and hole lifetimes of selected materials in broad temperature region 1.3 - 300 K, and photon echo signals detection on femtosecond time scale.

**3.1 Si-doped diamond films.** Diamond, with its hardness, high optical transmission in visible, and high thermoconductivity is a potentially useful material for multi-color hole-burning optical storage. Studies of chemical-vapor deposited (CVD) diamonds containing Si impurity centers with zero phonon line (ZPL) at 737 nm (1.6823 eV) were performed. The temperature range was 1.3 - 300 K. The two samples investigated were free-standing polycrystalline diamond films grown by microwave-assisted plasma CVD. Film #1 was spatially homogeneous, while film #2 had a nonuniform morphology related to growing conditions. The luminescence intensity of the sp<sup>2</sup> component, and intensity and width of the ZPL at 737 nm varied between films, and within the second film itself due to differences in morphology. Inhomogeneous broadening of ZPL was

determined of  $45\text{ cm}^{-1}$  for the film #1 and between of  $18$  and  $65\text{ cm}^{-1}$  for different spots on the film #2. At least six vibrational peaks were observed on the phonon sideband. The strongest line at  $515\text{ cm}^{-1}$  belongs to Si-Si bond and supports diatomic  $\text{Si}_2$  center structure. A decay time of  $950\text{ ps}$ , a luminescence quantum yield of  $0.04$ , and an absorption cross-section were determined for the Si center zero-phonon transition at  $77\text{ K}$ .

The first persistent spectral hole-burning of the Si impurity center in diamond films with ZPL at  $737\text{ nm}$ . was observed. Persistent spectral hole-burning was detected in the luminescence excitation spectra using tunable single-frequency Ti:sapphire laser at temperatures of  $1.3 - 20\text{ K}$ . Spectral holes with width of  $0.01\text{ cm}^{-1}$ , maximum depth up to  $30\%$ , and lifetime more than  $10^3\text{ sec}$  were observed between  $1.3$  and  $10\text{ K}$  ( $\Gamma_{\text{inh}}/\Gamma_{\text{hom}} = 5,000$ ). At higher temperatures holes became broader and shallower. Holes are resistant to temperature cycling: they persist even the sample is heated to at least  $120\text{ K}$ , and cooled back down to low temperatures. Hole width dependence on irradiation dose and temperature were studied. Processes of photoinduced hole filling by resonance light which compete with hole burning was discovered and studied. Those processes cause deleting of data while readout at temperatures above  $10\text{ K}$ . Using more pure material may help us to solve this problem. This material may be useful for writing and reading data at temperatures less than  $10\text{ K}$  and storage at  $77\text{ K}..$

**3.2 Diamond-like carbon and silicon-carbon allow films** Absorption and fluorescence spectra of diamond-like carbon and silicon-carbon allow films have been evaluated for their potential as hole-burning material. No useful ZPL were found.

**3.3 Rare-earth doped polymers and rare-earth doped sol-gel glasses** Absorption and fluorescence spectra of rare-earth doped polymers and rare-earth doped sol-gel glasses have been studied and evaluated for their potential as hole-burning material. The Eu<sup>3+</sup> and Tm<sup>3+</sup> organic complexes were synthesized by combining europium chloride and thulium nitrate with dibenzoylmethide, propionate, thenoyltrifluoroacetone and 2-hydroxy-5-methyl benzoate and ligands. These materials are potentially useful for high temperature hole-burning storage and their investigation will continue.

**3.4 Femtosecond photon echo.** In a parallel effort, time resolved research was performed in writing and retrieval of the femtosecond accumulated photon echo. A 2D imaging amplitude correlation technique was developed. This technique allows time to space domain conversion with femtosecond resolution. Four bit of information were stored in a hole-burning material (free-base octaethylporphyrin in polystyrene at 1.4 K) and retrieved by a single 150 fs pulse using CCD camera detection. Consequently, a read out speed of 27 Terabits per second has been demonstrated. Using this technique photon echo dependencies on exposure time, delay time between data and reference pulses, and number of readout cycles were studied. Up to 10<sup>4</sup> single-shot readout cycles by a femtosecond pulse were observed without substantial echo signal degradation. The echo decay time of 50 ± 15 ps was found for this sample.

#### **4. Presentations and publications.**

1. A. A. Gorokhovsky, G. Bai, and R. R. Alfano, "Ultrafast Excited States Dynamics of some Free-Base Porphyrins", 9th International Conference on the Dynamical Processes in Excited States of Solids, MIT, Cambridge, MA, August 4, 1993.

2. A. A. Gorokhovsky, A. V. Turukhin, R. R. Alfano, and W. Phillips, "Spectral hole-burning and fluorescence line narrowing of Si center in CVD diamond", *Spectral Hole-Burning and Related Spectroscopies: Science and Applications, Tokyo, Japan (August 24-26, 1994)*, 1994 OSA Technical Digest, Vol. 15, p. 240.
3. A. A. Gorokhovsky, A. V. Turukhin, R. R. Alfano, and W. Phillips, "Photoluminescence, fluorescence line narrowing, and spectral hole-burning studies of the Si center in CVD diamonds", presented at OSA Annual Meeting, Dallas, Texas, October 2-7, 1994, OSA Annual Meeting/ILS-X Program, ThBBB3 (1994).
4. I. Zeylikovich, G. Bai, A. Gorokhovsky, and R. R. Alfano, "Ultrafast retrieval of femtosecond photon echo signals", OSA Annual Meeting, Portland, OR, September 11, 1995.
5. A. V. Turukhin, A. Carpenter, C.-H. Liu, W. Sha, A. A. Gorokhovsky, R. R. Alfano, and W. Phillips, "Laser induced luminescence spectra and relaxation times of Si doped CVD diamonds", MRS 1995 Fall Meeting, Boston, MA, November 28, 1995.
6. A. A. Gorokhovsky, A. V. Turukhin, R. R. Alfano, and W. Phillips, "Persistent spectral hole burning of Si defects in CVD diamonds", MRS. 1995 Fall Meeting, Boston, MA, November 28, 1995.
7. A. A. Gorokhovsky, A. V. Turukhin, R. R. Alfano, and W. Phillips, "Photoluminescence vibration structure of Si center in chemical-vapor deposited diamond", *Appl. Phys. Lett.* **86**, 43 (1995).
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